## AMENDMENTS TO THE SPECIFICATION

*Please replace the paragraph beginning on Page 4, line 18 to read at follows:*

The sintered product 124 is heated, and the punch ~~124~~ 123 is downwardly moved. A compressive force is exerted on the sintered product 124. The plastic deformation takes place in the sintered product 124, and the sintered product 124 is stretched on the base plate 121 as shown in Figure 3B. The crystal grains of the sintered product 124 are oriented in a direction at which the figure of merit is improved. Thus, the thermoelectric semiconductor material 125 is improved in the figure of merit through the hot upset forging.

*Please replace the paragraph beginning on Page 7, line 12 to read at follows:*

Another problem is further encountered in the prior art crystal structure controlling technology described with reference to figures 3A and 3B in high electric resistivity. The ingot of solid solution of the thermoelectric material is pulverized into the powder before the pressure sintering. For this reason, the crystal grains of the sintered product 124 are relatively large and lack of uniformity. Even though the sintered product is subjected to the hot upset forging, the large and non-uniform crystal grains make the thermoelectric semiconductor 125 ~~exhibits~~ exhibit a large electric resistivity. In n-type thermoelectric semiconductor material, the large electric resistivity is serious.

*Please replace the paragraph beginning on Page 9, line 6 to read at follows:*

In accordance with yet another aspect of the present invention, there is provided a process for producing a thermoelectric material composed of at least one element selected from the group consisting of Bi and Sb and at least one element selected from the group consisting of Te and Se, and the process comprises the steps of a) preparing one of an ingot of

the thermoelectric material and a powder of the thermoelectric material, b) putting aforesaid one of the ingot and the powder into a die unit having an inlet portion and an outlet portion obliquely extending with respect to the inlet portion and c) pressurizing aforesaid one of the ingot and the powder for extruding a bulk of the thermoelectric material from the die unit at least once so that a ~~sharing~~ shearing force is exerted on aforesaid one of the ingot and the powder at a boundary between the inlet portion and the outlet portion.

*Please replace the paragraph beginning on Page 11, line 14 to read at follows:*

Fig. 9 is a ~~gram~~ graph showing a relation between the ratio of change in a power factor and temperature;

*Please replace the paragraph beginning on Page 13, line 17 to read at follows:*

The lamination is inserted into the inlet portion, and the punch exerts a force on the lamination. The lamination is pressed, and a ~~sharing~~ shearing force is exerted on the lamination at the boundary between the inlet portion and the outlet portion. The bulk 1 is extruded from the outlet portion. The extrusion is carried out once, or is repeated at least once. The direction in which the force is exerted on the lamination is hereinbelow referred to as "pressurization axis", and the direction in which the bulk 1 is extruded from the die unit is referred to as "extrusion axis". The pressurization axis and the extrusion axis are not coincident with one another in the die unit used in the process according to the present invention. The electric current is to flow in the direction substantially parallel to the extrusion axis.

*Please replace the paragraph beginning on Page 17, line 8 to read at follows:*

While a punch (not shown) is exerting a force on the lamination or powder in the inlet portion 2a, the lamination or powder is pressed against the inner surface defining the elbow portion of die unit 2, and the reaction from the inner surface acts as a ~~sharing~~ shearing force exerted on the lamination or powder. As a result, the lamination or powder is extruded from the inlet portion 2a into the outlet portion 2b, and the crystal grains are oriented through the rotation due to the ~~sharing~~ shearing force. When the bulk 1 is extruded from the die unit 2, most of the crystal grains have respective (001) planes substantially in parallel to or close to the extrusion axis C, and the average grain size is equal to or less than 30 microns. [001] direction is labeled with reference numeral 3, and θ2 is indicative of the angle between the [001] direction and the extrusion axis C, i.e., the inclination. The crystal grains with the inclination θ2 equal to or less than 45 degrees occupy an area not greater than 10 % of the total area of a surface 4 perpendicular to the extrusion axis C. In other words, most of the crystal grains on the surface 4 have the inclination greater than 45 degrees so that (001) planes thereof are in parallel to the extrusion axis C or cross the extrusion axis C at angle less than 45 degrees.

*Please replace the paragraph beginning on Page 16, line 3 to read at follows:*

The thermoelectric material thus produced through the above-described process has fine crystal grains. The average grain size is equal to or less than 30 microns. An inclination is defined as "angle between the [001] direction and the extrusion axis". The crystal grains with the inclination equal to or less than 45 degrees are determined on a surface cut ~~in~~ perpendicular to the extrusion axis by using an electron back scattering pattern. In detail, a sample of thermoelectric material is cut along a target surface, and the exposed surface is polished so as to create a smooth surface. While the smooth surface is being scanned by a scanning electron microscope, the distance or angle between the crossing points on the diffraction surface, which is observed with respect to the crystal structure, is measured, and the

direction of the planes exposed to the smooth surface is determined on the basis of the distance or angle.

*Please replace the paragraph beginning on Page 19, line 5 to read at follows:*

As will be understood from the foregoing description, the n-type thermoelectric material produced in accordance with the present invention is equivalent in thermoelectric properties to the p-type thermoelectric material. In general, the homogenous crystal orientation is required for n-type thermoelectric material, and the carrier concentration is precisely controlled for enhancing the Seebeck coefficient. Moreover, fine crystal grains are required for reduction in thermal conductivity. The starting material is obtained through the liquid quenching method so that the fine crystal grains and good carrier concentration control are achieved without adding any halogen. The ~~sharing~~ <u>shearing</u> force is exerted on the starting material in the die unit, which has the outlet portion not coincident with the inlet portion. While the ~~sharing~~ <u>shearing</u> force is being exerted on the material, the crystal grains are preferably oriented. This results in the n-type thermoelectric material equivalent in thermoelectric properties to the p-type thermoelectric material.

*Please replace the paragraph beginning on Page 20, line 6 to read at follows:*

A hydrogen reduction and sintering follows the liquid quenching. Thus, a sintered product is obtained through the sintering. A die unit formed with an inlet portion not coincident with an outlet portion is prepared. Any taper is not formed in the passage so that a piece of thermoelectric material is pressurized at only the boundary between the inlet portion and the outlet portion. The sintered product is inserted into the inlet port, and ~~is~~ a force is exerted on the sintered product with a punch. The sintered product is pressed against the inner surface at the boundary between the inlet portion and the outlet portion, and the reaction acts as a ~~sharing~~ <u>shearing</u> force. The sintered product is rotated at the boundary, and

a bulk of thermoelectric material is extruded from the outlet portion. The extrusion is carried out once, or is repeated at least once. After the extrusion, the thermoelectric material is treated with heat, and the bulk of the thermoelectric material is obtained.

*Please replace the paragraph beginning on Page 21, line 7 to read at follows:*

One of the particular features of the process according to the present invention is illustrated in figures 8A and 8B. In the process according to the present invention, the die unit is formed with the inlet portion 6a and the outlet portion 6b, and the centerline 7a of the inlet portion 6a crosses the centerline 7b of the outlet portion 6b. The ~~sharing~~ <u>shearing</u> force is exerted on the thermoelectric material at the boundary between the inlet portion 6a and the outlet portion 6b. As a result, the extruded bulk has the crystal grains with (001) planes 8a exposed to a surface 9a perpendicular to the extrusion axis 7b. In the prior art process, the centerline 7c of the inlet portion 6c is coincident with the centerline of the outlet portion 6d, and the crystal grains of the thermoelectric material are oriented during the sliding motion on the inner surface of the die unit. For this reason, [001] direction of each crystal grain is directed to the centerline 7d, and, accordingly, (001) planes 8b are arranged along the direction of the circumference on a surface 9b corresponding to the surface 9a. The crystal grains in the peripheral portion are strongly oriented, because the friction is directly exerted on the peripheral portion. However, the crystal grains in the central portion are less oriented, because the friction has been already consumed in the peripheral portion. Thus, the thermoelectric material produced through the process according to the present invention is more homogenous than the thermoelectric material produced through the prior art process. This results in that the production yield is enhanced by virtue of the process according to the present invention.

*Please replace the paragraph beginning on Page 22, line 7 to read at follows:*

The present inventors investigated the influence of the composition ratio between Te and Se on the thermal properties. The thermoelectric material is expressed as (Bi, Sb)2 (Te, Se)3. The present inventors produced samples of thermoelectric material different in composition ratio between Te and Se. Using the samples, the present inventors fabricated thermoelectric modules on substrates, and electric current ~~flew~~ flow through the thermoelectric modules. The present inventors measured the temperature characteristics between -20 degrees to 100 degrees in centigrade, and calculated a ratio of change in power factor. When the thermoelectric modules were operated at room temperature, i.e., 25 degrees in centigrade, the power factor is 1. The ~~radio~~ ratio of change in power factor was compared among the samples, and the present inventors confirmed that the composition ratio Te/ Se between 2.5/ 0.5 and 2.7/ 0.3 made the ratio of change in power factor highest as shown in figure 9.

*Please replace the paragraph beginning on Page 22, line 20 to read at follows:*

Description is hereinbelow made on process parameters for producing the thermoelectric material ~~fallen within the technical scope of the present invention~~ according to the present invention.

*Please replace the paragraph beginning on Page 23, line 1 to read at follows:*

As described hereinbefore, the die unit used for the extrusion has the inlet portion and the outlet portion not coincident with one another, and the ~~sharing~~ shearing force is exerted on the thermoelectric material at the boundary between the inlet portion and the outlet portion. The angle between the pressurization axis and the extrusion axis and the temperature of the thermoelectric material in the extrusion have strong influences on the crystal structure of the thermoelectric material.

*Please replace the paragraph beginning on Page 23, line 8 to read at follows:*

The present inventors investigated the influences of these two factors on the crystal structure. The present inventors prepared the die units different in the angle between the pressurization axis and the extrusion axis and plural kinds of thermoelectric material with the composition expressed as $(Bi, Sb)_2 (Te, Se)_3$. Any taper was not formed in the passages in the die units. The ratio between Te and Se was ~~fallen~~ within the above-described range. The present inventors extruded bulks of the thermoelectric material from those die units, and observed the crystal structure. The present inventors further evaluated the extruded bulks of the thermoelectric material from the viewpoint of the thermoelectric properties.

*Please replace the paragraph beginning on Page 27, line 4 to read at follows:*

The present inventors calculated the relative density of samples formed of $Bi_{1.9}Sb_{0.1}Te_{2.6}Se_{0.4}$, and <u>in Figure 11</u> plotted the values in terms of the extrusion ratio. The relative density was representative of the ratio between the density of the sample and the density of a reference sample. The reference sample was the thermoelectric material expressed as $Bi_2Te_3$ , which was 7.858 grams/ $cm^3$ as written in ASTM (American Society of Testing Method) card. In other words, the relative density of the reference sample was 100 % .

*Please replace the paragraph beginning on Page 29, line 18 to read at follows:*

The present inventors further investigated the influence of repetition on the figure of merit. The present inventors prepared samples formed of the thermoelectric material. The present inventors repeatedly extruded the samples from a die unit. The die unit had the angle between the pressurization axis and the extrusion axis adjusted to 90 degrees, and the extrusion was carried out in argon at 450 degrees in centigrade. The punch was moved at 0.03 millimeter per minute. However, the number of times repeated was different among the samples. The present inventors evaluated the samples from the viewpoint of the figure of

merit. The present inventors firstly confirmed that the bulk of thermoelectric material subjected to the extrusion at least once was larger in figure of merit than the bulk of thermoelectric material subjected to the extrusion only once. The present inventors further confirmed that the maximum ~~share~~ shear stress was increased proportional to the number of times repeated and that the average grain size was reduced inversely proportional to the number of times repeated.

*Please replace the paragraph beginning on Page 30, line 10 to read at follows:*

Figure 13 shows the relation between the number of times repeated and the average grain size and the relation between the number of times repeated and the maximum share stress. Plots "x" were representative of the relation between the number of times repeated and the maximum ~~share~~ shear stress in the samples, and dots stood for the relation between the number of times repeated and the average grain size of the samples. When the number of times was increased, the average grain size was reduced. On the other hand, the maximum ~~share~~ shear stress was increased together with the number of times repeated.

*Please replace the paragraph beginning on Page 30, line 22 to read at follows:*

The present inventors further investigated influences of the extrusion speed on the thermoelectric properties. The present inventors prepared samples of the thermoelectric material, and extruded the samples from a die unit in the technical scope of the present invention at different extrusion speed. The present inventors calculated the figure of merit, and found that the extrusion speed was to be ~~fallen~~ within the range from 0.01 millimeter per minute to 1 millimeter per minute. The present inventors further confirmed that the extrusion speed between 0.05 mm/ min. and 0.2 mm/ min. was more preferable.

*Please replace the paragraph beginning on Page 32, line 9 to read at follows:*

After the extrusion, the thermoelectric material according to the present invention is subjected to a post treatment in a direction parallel to the extrusion axis on the plane defined by the pressurization axis and the extrusion axis as follows. One of the post treatments is an SPS (Spark Plasma Sintering). Another post treatment is a hot pressing by using a forging machine. The present inventors investigated influences of the post treatment on the thermoelectric properties. The present inventors prepared samples of the thermoelectric material, and divided the samples into three groups. The samples were formed of thermoelectric material expressed as $Bi_{1.9}Sb_{0.1}Te_{2.6}Se_{0.4}$. The samples of the first group were produced from the flakes obtained through the liquid quenching method identical in conditions to those of the second embodiment, and were subjected to the hot pressing without any extrusion. The samples of the second group were produced through the process implementing the second embodiment. The samples of the second group were subjected to the extrusion. The die unit had the inlet portion not coincident with the outlet portion, and the angle between the pressurization axis and the extrusion axis was 90 degrees. The extrusion was carried out at 450 degrees in centigrade at 0.1 mm/ min. However, the samples of the second group were not subjected to the hot pressing. The samples of the third group were subjected to the hot pressing after the extrusion. The present inventors measured the electric resistivity, and determined the Seebeck coefficient. The present inventors plotted the thermoelectric properties of the samples in figure 14. A ~~bubble~~ circle stood for one of the samples in the first group, triangle was representative of the relation observed in the samples of the second group, and x stood for one of the sample in the third group. Three linear lines represented the power factor P.F. of $3.0 \times 10^{-3}$ W/(K$^2$m), $3.5 \times 10^{-3}$ W/(K$^2$m) and $4.0 \times 10^{-3}$ W/(K$^2$m).

*Please replace the paragraph beginning on Page 33, line 20 to read at follows:*

From figure 14, the samples of the first group were large in both of the Seebeck coefficient and the electric resistivity, because the crystal grains were not strongly oriented

through the hot pressing. As a result, the power factor P.F. was close to the linear line representing $3.0 \times 10^{-3}$ W/($K^2$m). The samples of the second group and the samples of the third group were subjected to the extrusion, and the electric resistivity was lower than that of the samples of the first group. The samples of the second group were at the middle point between the leaner lines representing $3.0 \times 10^{-3}$ W/($K^2$m) and $3.5 \times 10^{-3}$ W/($K^2$m). On the other hand, the samples of the third group were ~~fallen~~ within the range between $3.5 \times 10^{-3}$ W/($K^2$m) and $4.0 \times 10^{-3}$ W/($K^2$m). Comparing the triangle with the mark "x", it was understood that the hot pressing after the extrusion was conducive to the improvement of the thermoelectric properties. When the samples were hot pressed in the direction perpendicular to the extrusion axis on the virtual plane defined by the pressurization axis and the extrusion axis, the crystal grains became finer without changing the orientation thereof.

*Please replace the paragraph beginning on Page 36, line 7 to read at follows:*

The present inventors investigated influences of the hydrogen reduction on the thermoelectric properties. The present inventors prepared samples of the thermoelectric material, and divided the samples into two groups. The ~~staring~~ <u>starting</u> material for all the samples was the flakes produced through the liquid quenching method. The samples of the first group were treated in the hydrogen reduction at 400 degrees in centigrade, and, thereafter, the samples were extruded from the die unit. On the other hand, the samples of the second group were extruded from the die unit without the hydrogen reduction. The hot pressing was not carried out for the samples.

*Please replace the paragraph beginning on Page 37, line 22 to read at follows:*

~~Figure 14~~ <u>Figure 15</u> shows the preferable pressure applied to the samples of the first group and the samples of the second group. The preferable pressure to be applied to the samples of the first group was 9.31 kN/ $cm^2$, i.e., 0.95 ton-weight/ $cm^2$. The preferable

pressure to be applied to the samples of the second group was 11.47 kN/ $cm^2$, i.e., 1.17 ton-weight/ $cm^2$. Thus, the present inventors confirmed that the lamination was preferable from the viewpoint of reduction in pressure.

*Please replace the paragraph beginning on Page 39, line 15 to read at follows:*

The sintered product is put into a die unit which has an inlet portion not coincident with an outlet portion. The sintered product is pressurized in the die unit, and a ~~sharing~~ <u>shearing</u> force is exerted on the sintered product during the extrusion. The extrusion is carried out once, or is repeated at least once. The extruded body is treated with heat. Then, a bulk of thermoelectric material is obtained through the process implementing the third embodiment. The average grain size is equal to or less than 30 microns, and the crystal grains, which have respective [001] directions crossing the extrusion axis at 45 degrees or less, occupy an area equal to or less than 10 percent of the cross section perpendicular to the extrusion axis.

*Please replace the paragraph beginning on Page 42, line 6 to read at follows:*

The thermoelectric elements 121a/ 121b are prepared as follows. A bulk of n-type thermoelectric material and a bulk of p-type thermoelectric material are produced through the process implementing any one of the first to third embodiments as by step S11. The bulk of p type thermoelectric material and the bulk of n-type thermoelectric material are sliced as by step S12. Subsequently, nickel is plated on the slices through an electroless plating technique as by step S13. Nickel and gold may be plated through the electroless plating technique. Finally, ~~he~~ <u>the</u> slices are separated into dices as by step S14. Thus, the thermoelectric elements 121a/ 121b are produced through the steps S11, S12, S13 and S14.

*Please replace the paragraph beginning on Page 43, line 11 to read at follows:*

The present inventor evaluated the Peltier module 120. The present inventors fabricated samples of the Peltier module through the process shown in figure 18 and samples of the prior art Peltier module through a corresponding process. The prior art p/ n type thermoelectric elements were incorporated in the samples of the prior art Peltier module. The present inventors ~~flew~~ applied an electric current through the samples of the present invention as well as the samples of the prior art module. Temperature difference was adjusted to a predetermined value. The present inventors measured the electric power consumption. The present inventors confirmed that the electric power consumption was reduced at 20 % as shown in figure 19.

*Please replace the paragraph beginning on Page 46, line 3 to read at follows:*

The present inventors produced comparative samples. Pellet No. 11 and pellet No. 12 were comparative samples. These pellets were subjected to hot pressing instead of the extrusion. For this reason, any ~~sharing~~ shearing force was not exerted on those pellets.

*Please replace the paragraph beginning on Page 46, line 5 to read at follows:*

Table 5 shows the composition of the pellets and the conditions in the process. When the extrusion was repeated at least once, table 5 shows the extrusion ratio in the last extrusion. Pellet Nos. 1 to 10 were ~~fallen~~ within the technical scope of the present invention, and pellet Nos. 11 and 12 were comparative samples. In table 5, abbreviations "AG", "TM", "RT", "TP" and "SP" stand for the angle between the pressurization axis and the extrusion axis, the number of times repeated, the extrusion ratio, the temperature during the extrusion and the extrusion speed.

*Please replace the paragraph beginning on Page 47, line 1 to read at follows:*

The present inventors evaluated the pellets. The Seebeck coefficient $\alpha$, electric resistivity $\rho$ and thermal conductivity $\kappa$ were measured, and the figure of merit was calculated therefrom. The present inventors further measured the crystal grains with [001] direction, i.e., c-axis inclined to the extrusion axis by 45 degrees or less through the EBSP (Electronic Back-Scatter Pattern), and determined the ratio of the area occupied by the crystal grains to the total area of a cross section perpendicular to the extrusion axis. The present inventors further analyzed the cross section perpendicular to the extrusion axis through the x-ray diffraction, and determined the ratio between diffraction intensity I(100) representative of (110) plane to the diffraction intensity I(015) representative of (015) plane, i.e., I(110)/ I(015). The results were summarized in table 6. In table 6, "Seebeck" means Seebeck Coefficient, abbreviations "R", "OF", "TC", "FM", "R1" and "R2" stand for the resistivity, the power factor, the thermal conductivity, the figure of merit, the ratio I(110)/ I(015) and the ratio of the area occupied by the crystal grains to the total area of the cross section.

*Please replace the paragraph beginning on Page 49, line 19 to read at follows:*

Figure 20 shows a relation between the dispersion ratio and the angle θ2 between [001] direction and the extrusion axis. Dots stood for sample No. 2. ~~Bubbles~~ Circles were representative of another sample which was produced under the same conditions except for the temperature during the extrusion. Although sample No. 2 was extruded at 450 degrees in centigrade, the other sample was extruded at 380 degrees in centigrade.

*Please replace the paragraph beginning on Page 50, line 18 to read at follows:*

The p-type thermoelectric material and the n-type thermoelectric material and the metal form the p-type thermoelectric elements and the n-type thermoelectric elements, and the p-type thermoelectric elements and the n-type thermoelectric elements are alternately connected in series on the insulating substrates. ~~Since the~~ The p-type/ n-type thermoelectric material is large in figure of merit so that the power consumption of the thermoelectric module is surely reduced.